AFRL-SR-AR-TR-02-0173

FINAL REPORT FOR PERIOD JAN-DEC 2001
AFOSR GRANT: F49620-96-1-0092

F49620-96-1-0092

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20020614 175

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Air Force Office of Scientific Research
Arlington, Virginia

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REPORT DOCUMENTATION PAGE

AFRL-SR-AR-TR-02-
0173

1. REPORT DATE *(DD-MM-YYYY)* 01-02-02	**2. REPORT TYPE** FINAL	**3. DATES COVERED** *(From - To)* JAN DEC 2001
4. TITLE AND SUBTITLE FINAL REPORT FOR PERIOD JAN-DEC 2001 AFOSR GRANT: F49620-96-1-0092		**5a. CONTRACT NUMBER**
		5b. GRANT NUMBER F49620-96-1-0092
		5c. PROGRAM ELEMENT NUMBER 61102F
6. AUTHOR(S) Philp D. Whitefield Donald E. Hagen		**5d. PROJECT NUMBER** 2303
		5e. TASK NUMBER BX
		5f. WORK UNIT NUMBER
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) UMR CASL G-7 Norwood Hall UMR Rolla, MO 65401		**8. PERFORMING ORGANIZATION REPORT NUMBER**
9. SPONSORING / MONITORING AGENCY NAME(S) AND ADDRESS(ES) AFOSR		**10. SPONSOR/MONITOR'S ACRONYM(S)**
		11. SPONSOR/MONITOR'S REPORT NUMBER(S)

12. DISTRIBUTION / AVAILABILITY STATEMENT
N/A

13. SUPPLEMENTARY NOTES
N/A

14. ABSTRACT
This report describes work performed under AFOSR contract F49620-96-1-0092 during the calendar year 2001. The major effort during this period has been data analysis, interpretation and publication of results acquired in this research program. To data there have been two presentations at peer-reviewed symposia and one publication submitted to peer-reviewed journals. Two additional papers are under preparation for submission to the journals Aerosol Science and Technology and the Journal of Geophysical Research. The publications from this effort are attached as appendices and represent the progress achieved during the period of performance.

15. SUBJECT TERMS

16. SECURITY CLASSIFICATION OF:			**17. LIMITATION OF ABSTRACT**	**18. NUMBER OF PAGES**	**19a. NAME OF RESPONSIBLE PERSON** Philip D. Whitefield
a. REPORT	**b. ABSTRACT**	**c. THIS PAGE**			**19b. TELEPHONE NUMBER** *(include area code)* 573-341-4340

PROJECT SUMMARY

In-situ measurements of the total aerosol particle concentration (370*nm* to 4000*nm*) and differential particle size distribution (8*nm* to 4000*nm*) were made in the upper tropospheric/ lower stratospheric sections of several rocket exhaust plumes. The rockets that created these exhaust plumes included the Atlas IIAS, Delta II, Athena II, SST and Titan . The measurements were made using Differential Mobility Analysis (DMA) and Laser Particle Counting (LPC) techniques. The research aircraft used to make the stratospheric plume incursions was a WB-57F. The aerosol sampling instrument that was flown on the WB-57F was called the ACCENT Mobile Aerosol Sampling System (ACCENTMASS) and combined a Grab Tank Sampling (GTS) system and a Real-time Particle Measurement System (RPM). The former was used in conjugation with Differential Mobility Analysis (DMA) after the WB-57F had landed, whilst the latter used Laser Particle Counters (LPC) to collect data continuously throughout the flight. The data presented in this thesis were measured during the Atmospheric Chemistry of Combustion Emissions Near to the Tropopause (ACCENT) mission.

The total aerosol particle concentration data (370*nm* to 4000*nm*) were measured using the Real-time Particle Measurement System (RPM), , as temporal total concentration profiles for the whole aerosol particles and the non-volatile aerosol particles (available only for the Athena II). The total aerosol particle concentration falls from between 750±100 particles/cm^3 to between 350±100 particles/cm^3 over 2000 seconds, which corresponds to an approximate plume dispersion rate of 0.175 to 0.200 particles/($cm^3 \cdot s$)

The frequency of the total aerosol particle concentration measurement (370*nm* to 4000*nm*) was 1.67 Hz, which was high enough to reveal detailed structure in the plume. Furthermore, the signal to noise ratio in the plume was at least 100:1 in the total aerosol particle concentration measurements, which allowed the WB-57F Flight Engineer to rely on the RPM system to detect the edge of the plume when making an incursion.

The number based differential particle size distributions (dN/dx) were measured between 8*nm* and 4000*nm* using Differential Mobility Analysis (DMA) (8*nm* to 250*nm*) and Laser Particle Counting techniques (370*nm* to 4000*nm*). The size distributions were trimodal. The concentration maxima of Mode 1 fell below the detection window of the ACCENTMASS (<8*nm*). The concentration maxima of Modes 2 and 3 are clearly observed. The diameters of maximum concentration of Modes 1, 2 and 3 were <10*nm*, 50*nm* to 60*nm*, and 915*nm* to 983*nm* respectively. These results compare to within 50 percent in most cases with data collected previously by other investigators Some investigators claim the presence of a mode above 2500*nm*. No evidence of such a mode was seen in the Atlas IIAS, Delta II or Athena II data, but the ACCENTMASS is not designed to measure aerosol containing particles larger than 4000*nm*.

The diameters at which these three modes peaked in concentration were not statistically different between the rockets measured during the ACCENT missions, indicating that from the UMR data no significant relationship can be seen between the exhaust particle size in the range of motor sizes or types used by these rockets. However, the average diameter of maximum differential concentration in Mode 2 may increase as the mass flow from the Solid Rocket Motor (SRM) contributes more greatly to the total mass flow from a rocket that contains either both kerosene-liquid oxygen (RP/LOx) and SRMs (Atlas IIAS and Delta II), or just an SRM (Athena II). Such a relationship would imply that the SRM produces larger Mode 2 particles (~60*nm*) than the RP/LOx motor (~50*nm*). The size of the SRM used by each rocket increases in the following order: Atlas IIAS <DeltaII <Athena II (Lockheed Martin Website, 2002). Also, as the size of the SRM increases, the diameter of maximum differential concentration may also increase. The analyses performed on the diameters of maximum concentration imply, but do not state conclusively that fuel type and SRM motor size both affect the diameter of maximum concentration in Mode 2.

The ratio between average maximum concentrations of Modes 2 and 3 and the ratio of mass flows from RP/LOx and SRM motors were related. The mathematical expression that describes this relationship is $y = 799.79e^{-1.1539x}$. The relationship

indicates that the SRM motors generate more Mode 2 particles than the RP/LOx motors. In the Atlas IIAS, the Delta II and the Athena II, no significant difference was seen in the quantity of Mode 3 particles generated by either motor type.

Good agreement exists between the particle size distributions measured with the GTS and RPM systems. The agreement was measured as the magnitude of the discontinuity where the particle size distributions were coupled. The discontinuity magnitude was referenced to the difference between the concentration maxima of Modes 2 and 3. The average magnitude of the discontinuity is 1.12 ± 1.06 percent of the difference between the concentration maxima of Modes 2 and 3. Such good agreement justifies the coupling of the dN/dx particle size distributions produced by the DMA and LPC techniques.

Mode 3 (370*nm* to 4000*nm*) whole aerosol particle dN/dx size distribution was not seen to vary as the WB-57F traversed the Athena II plume during plume incursions 1, 4 and 6. As afterburning and other plume processes are very turbulent, by the time the WB-57F has first entered the plume any regional variation in the particle size distribution may have disappeared.

The differences between the integrals of the Mode 3 whole aerosol and non-volatile aerosol volume based differential particle size distributions (dV/dx) were calculated for every plume incursion in the Athena II flight. The average of these differences was interpreted as the average volume of a volatile component made of ice, nitric acid trihydrate and various chlorine species that evaporated from the particle surface in the thermal discriminator. The average volume of volatile material was calculated to be 43 ± 15 percent of the whole aerosol particle volume. The apparent decrease in average diameter of maximum differential concentration from 983*nm* to 916*nm* as the aerosol passes through the discriminator supports the presence of a volatile component in the aerosol particles. However, this apparent reduction in particle diameter is not statistically significant when the uncertainty in the Laser Particle Counter (LPC) sizing is considered, and may be partially caused by changes in the refractive index of the particle. The data suggest that the refractive index of the alumina aerosol particles is modified when they are covered by this surface layer of volatile material. The resulting

change in the scattering properties that occurs when the particle passes through the thermal discriminator may cause the particle to be over- or under-sized by LPC1 (the whole aerosol LPC). The error in sizing that is caused by the change of particle refractive index when the volatile surface layer is gained, influences the calculation of the volume of volatile component. Therefore the statistical significance and quantity of the volume of the volatile component needs to be verified by future work.

The aerosol particle size distributions measured during this research project support the conclusions made by Jackman, *et al.*, (1998) and Danilin, *et al.*, (2001), which state that the effect of rocket exhaust aerosol on regional and global stratospheric ozone concentrations will be small.